

Train of high-power femtosecond pulses: Probe wave in a gas of prepared atoms

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Abstract

We present a new method for generating a regular train of ultrashort optical pulses in a prepared two-level medium. The train develops from incident monochromatic probe radiation travelling in a medium of atoms, which are in a quantum mechanical superposition of dressed internal states. In the frame of linear theory for the probe radiation, the energy of individual pulses is an exponentially growing function of atom density and of interaction cross section. Pulse repetition rate is determined by the pump field's generalized Rabi frequency and can be around 1 THz and greater.

We also show that the terms, extra to the dipole approximation, endow the gas by a new property: non-saturating dependence of refractive index on dressing monochromatic field intensity. Contribution of these nonsaturating terms can be compatible with the main dipole approximation term contribution in the wavelength region of about ten micrometers (the range of CO_2 laser) or larger.

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Quantum interference is one of principal values of quantum theory that essentially enriches the list of earlier ordered principles of physical theory. Now there is a good comprehension how to prepare the system in a desirable quantum state of coherent superposition of two or more eigenstates of a quantum system. This is especially relevant to manipulations with a laser radiation where some new paths for scientific and technological applications have been established due to extensive theoretical and experimental studies in the last two-three decades, e.g. quantum information processing [1], lasing without inversion [2].

The scenario discussed in this paper, where a superposition of two dressed atomic states is coupled to a weak (probe) quasimonochromatic laser radiation also takes advantage of just this quantum interference effect. Of particular pertinence, to the subject under consideration, is the work of S.E. Harris and co-workers, who have suggested and used a Raman-type three level interaction scheme in D_2 -molecular gas to get series of femtosecond pulses [3]. The applied lasers, driving the molecular Raman transition slightly off-resonance, cause the molecules to vibrate in unison. Time-varying refractive index of such a non stationary gas generates ultrabroad spectrum of Raman sidebands whose Fourier transform may synthesize a train of desired ultrashort pulses. The usage of quantum interference effects in order to manipulate the optical properties of gaseous atomic or molecular medium has by now been established as a useful and powerful method. In particular, by means of destructive interference of two coupled (Raman) optical transitions, a resonant, opaque medium can be made transparent for radiation inducing these transitions [4]. This is the well-known phenomenon of electromagnetically induced transparency (EIT) [5], where the vanishing absorption is accompanied by an enhanced nonlinear response [6]. It sets up the carrier idea for a number of promising suggestions and applications, such as, slow light [7] and light stopping [8] in driven atomic systems. Note also, that the quantum superposition principle takes off any restriction on the amount of momentum transferred from one energy level to another during a single-photon absorption/emission process [9].

In this paper we suggest and analyze a new theoretical model for producing a train of ultrashort optical pulses. We show that an incident quasimonochromatic radiation (probe wave) attains a periodic modulation of intensity accompanied, in general, by amplification, if the medium atoms have been prepared in a quantum superposition state of two internal states, dressed by the pump field. For appropriate gas parameters, concentration and resonance detuning, the probe wave intensity modulates in a form of regular train of sharp ultrashort pulses. Distance between these pulses is determined only by the Rabi frequency of the pump field, while duration and intensity of each pulse are determined by other parameters also, e.g. by the product of atomic concentration on interaction cross section.

So we consider a gas of two-level atoms (with energy difference $\hbar\omega_0$ between the excited and ground internal atomic bare states $|2\rangle$ and $|1\rangle$) in a far off-resonance field of monochromatic radiation, regarded as the pump field. The spin of relevant to optical transition electron and the possible sublevel structures, as well as the frequency Doppler shifts will not

be taken into account. The atomic(internal) state in dressed state basis is given by [10]:

$$\Psi_{\pm} = N_{\pm} \left(|1\rangle - \frac{2\lambda_{\pm}}{\Omega} |2\rangle e^{-i\omega_p t + ik_p z} \right) \times \exp \left(-\frac{i}{\hbar} (E_1 + \hbar\lambda_{\pm}) t \right). \quad (1)$$

The normalization coefficient $N_{\pm} = \Omega / \sqrt{2\Omega'(\Omega' \mp \Delta)}$, where $\Omega = 2dE_p/\hbar$ and $\Omega' = \sqrt{\Delta^2 + \Omega^2}$ are the normal and generalized Rabi frequencies in the pump field, $\lambda_{\pm} = -\Delta/2 \pm \Omega'/2$ is the high-frequency Stark shift of energy levels for positive (+) or negative (-) detuning $\Delta = \omega_p - \omega_0$, and E_1 is the energy of the bare state $|1\rangle$. In fact the detuning Δ is assumed much larger than the full linewidth of the optical transition, and thus spontaneous emission and other inelastic processes are strongly suppressed and can be neglected for timescales characteristic for observing the later described effects.

Propagation of a weak probe radiation through a system of dressed atoms can be described by a wave equation

$$\left(\vec{\nabla}^2 - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) \vec{A}_{probe}(\vec{r}, t) = -\frac{4\pi\rho}{c} \frac{\partial}{\partial t} \langle \hat{d} \rangle_{probe}, \quad (2)$$

for a probe field vector potential $\vec{A}_{probe}(\vec{r}, t) = \vec{A}(\vec{r}, t) \exp[ik\vec{r} - i\omega t] + c.c.$ with slowly varying amplitude $\vec{A}(\vec{r}, t)$. ρ is the atom number density and $\langle \hat{d} \rangle_{probe}$ is the atomic dipole moment induced by a probe field. To acquire the latter, one has to find first the atomic state $|\Psi(\vec{r}, t)\rangle$ in a combined field of dressing and probe fields, then implement the ordinary quantum mechanical averaging of the dipole operator by means of this state vector, and later select terms proportional to the perturbing field $\vec{A}(\vec{r}, t)$. It should be added here that the right side of Eq.(2) examines the case of isolated atoms, or atoms in a rather dilute gas, where collisional relaxation and dephasing effects can be ignored.

The atomic state vector has the form

$$|\Psi(\vec{r}, t)\rangle = (\alpha + C_+(\vec{r}, t)) |\Psi_+(z, t)\rangle + (\beta + C_-(\vec{r}, t)) |\Psi_-(z, t)\rangle, \quad (3)$$

and correspondingly

$$\begin{aligned} \langle \hat{d} \rangle_{probe} = & (\alpha^* C_+ + \alpha C_+^*) \langle \Psi_+ | \widehat{\vec{d}} | \Psi_+ \rangle + \\ & (\alpha^* C_- + \beta C_+^*) \langle \Psi_+ | \widehat{\vec{d}} | \Psi_- \rangle + \\ & (\alpha C_-^* + \beta^* C_+) \langle \Psi_- | \widehat{\vec{d}} | \Psi_+ \rangle + \\ & (\beta^* C_- + \beta C_-^*) \langle \Psi_- | \widehat{\vec{d}} | \Psi_- \rangle. \end{aligned} \quad (4)$$

where the complex numbers α and β (we assume them to be known in following discussion) represent the probability amplitudes of dressed states and constitute the mentioned superposition state for the atom in a field of pump laser radiation. Such states can be generated, for example, by using sufficiently short times for raising the pump field intensity from zero to the atomic state dressing value. This rapid ramp, eliminating the condition of adiabaticity,

creates for the atom a superposition of two dressed states. Nonvanishing population of both states requires a switching on time, the inverse value of which is not much smaller than the frequency detuning. For instance, if $\Delta = 2 * 10^{11} \text{ Hz}$, a switching on time of about 10 psec is needed to get perceptible percents in the states superposition. Similar results may be obtained also by means of well-established technique of Stark switching on of interaction[11].

The additional terms $C_+(\vec{r}, t)$ and $C_-(\vec{r}, t)$ are perturbations due to interaction with probe radiation which are proportional to the probe field vector potential amplitude $\vec{A}_{probe}(\vec{r}, t)$. They have to be determined from the Schrödinger equation yet. Standard calculations yield

$$i\hbar\partial C_{\pm}/\partial t = \alpha \langle \Psi_{\pm} | \hat{V}_{probe} | \Psi_{+} \rangle + \beta \langle \Psi_{\pm} | \hat{V}_{probe} | \Psi_{-} \rangle, \quad (5)$$

where

$$\begin{aligned} \hat{V}_{probe}(\vec{r}_e, t) = & -\frac{e}{c} \vec{A}_{probe}(\vec{r}_e, t) \hat{\vec{p}} - \\ & \frac{e^2}{mc^2} \vec{A}_p(z_e, t) \vec{A}_{probe}(\vec{r}_e, t) \end{aligned} \quad (6)$$

is the probe's interaction energy and is obtained from free atom Hamiltonian after common substitution $\hat{\vec{p}} \rightarrow \hat{\vec{p}} - (e/c) \vec{A}_T(\vec{r}, t)$ (here $\vec{A}_T(\vec{r}, t)$ is the total, pump + probe vector potential) and selection of terms proportional to the weak probe field vector potential. Note, the ordinary dipole approximation is represented by the first term in Eq. (6), hence our analysis extends beyond the dipole approximation for the probe field.

Calculation of matrix elements in equations (5) supposes integration over the optical electron coordinate relative to the atomic center of mass (a.c.m.), and thus we introduced $\vec{r}_e = \vec{r} + \vec{\xi}$, \vec{r} being the a.c.m. radius vector. Here we suppose, that the bare atomic states have no permanent dipole moment. Due to opposite spatial parities this immediately exterminates also the bare state nondiagonal matrix elements $\langle 1 | \dots | 2 \rangle$, $\langle 2 | \dots | 1 \rangle$ of the out of dipole approximation term of Hamiltonian (6). Insertion of found solutions of Equations (5) into (4) determines the right hand side of wave equation (2) as an explicit function of system parameters, proportional to the probe wave vector potential $\vec{A}_{probe}(\vec{r}, t)$. In the next step we apply to this general form wave equation the approximation of slowly varying coefficients and attain the reduced wave equation (r.w.e.) for the slowly varying amplitude $\vec{A}(\vec{r}, t)$. Some of the right-hand side terms of obtained r.w.e. are responsible for parametric generation processes, parametric down conversion and 4-wave parametric amplification respectively. However in this paper these processes will not be considered and we will restrict ourselves with a more simple non-parametric propagation. In familiar formulation this approach, as is well known, leads to determination of the medium refractive index.

The terms which describe the medium optical properties can now be divided into two groups: one which contains a propagation-type exponential factor $\exp(i \vec{k} \vec{r} - i\omega t)$ and the second group, where the frequency in exponential factors is red- or blue-sideband shifted by a relatively small amount $\Omega' = \sqrt{\Delta^2 + \Omega^2}$. They represent a bias current on incident probe wave frequency ω and on two frequencies symmetrically shifted from ω by the generalized Rabi frequency Ω' . Existence of these sideband components temporarily modulates the

bias current. The light travelling through the medium perceives it as a time varying dielectric constant and becomes modulated in frequency as well as in amplitude. In following discussion we are analyzing consequences of this modulation.

So, the r.w.e. can be written in the following symbolic form:

$$\left(\vec{n} \frac{\partial}{\partial \vec{r}} + \frac{1}{c} \frac{\partial}{\partial t} \right) A(\vec{r}, t) = i(D + LS \exp(-i\Omega') + RS \exp(i\Omega')) A(\vec{r}, t) \quad (7)$$

where $\vec{n} = \vec{k}/k$ is a unit vector in probe field propagation direction, in general distinct from the pump field direction. D , LS and RS (together with $A(\vec{r}, t)$) give the mentioned direct, red- and blue- sideband components of bias current in a two-level dielectric gas medium. The first one of them is proportional to population difference $|\alpha|^2 - |\beta|^2$ and yields the familiar index of refraction (see Eq.(9)). It has no conceptual relevance to the superposition nature of the medium, as we would obtain the same result for a gas medium a unit volume of which contains $|\alpha|^2 \rho$ atoms in the ground state and $|\beta|^2 \rho$ atoms in the excited one. Superposition nature exhibits itself only in the red- and blue-sideband components, which are proportional to the product of the probability amplitudes α and β (see Eq.(10)). It is of special importance for the following results that though LS and RS are real, in general $LS \neq RS$ and the contribution of red- and blue-sidebands has real and imaginary parts.

To solve Eq.(7) we assume as a boundary condition that the probe field is applied perpendicular at the boundary of the medium and is of infinite extent in transverse direction. Now (taking into account also the explicit expressions of D , LS and RS) the seeking solution for the probe wave vector potential at $t - (\vec{n} \vec{r})/c \geq 0$ has a form

$$A_{probe}(\vec{r}, t) = A_0 F(\vec{r}, t) e^{i\omega n_0(\omega) \vec{k} \vec{r}/c - i\omega t} + c.c., \quad (8)$$

where A_0 is the input wave amplitude,

$$n_0(\omega) = 1 + \frac{\pi \rho}{2\omega^2} (|\alpha|^2 - |\beta|^2) \times \left(\left(\frac{d^2 \omega_0^2 (\Omega' - \Delta)^2}{\hbar \Omega'^2} + \frac{e^2 \Omega'^2}{m \Omega'} \right) \frac{1}{\omega_p - \omega + \Omega'} - \left(\frac{d^2 \omega_0^2 (\Omega' + \Delta)^2}{\hbar \Omega'^2} + \frac{e^2 \Omega'^2}{m \Omega'} \right) \frac{1}{\omega_p - \omega - \Omega'} \right) \quad (9)$$

is the ordinary refractive index and the prefactor

$$F(\vec{r}, t) = \exp \left(\frac{2\pi \rho d^2 \omega_0^2 \Omega}{\hbar \omega \Omega'^3} (f_1 - f_2) \right) \quad (10)$$

with

$$f_1 = \frac{\alpha^* \beta (1 - \exp(-i\Omega' (\vec{n} \vec{r})/c))}{\exp(i\Omega' t) \left(\frac{\Omega' + \Delta}{\omega_p - \omega} + \frac{\Omega' - \Delta}{\omega_p - \omega + \Omega'} \right)}$$

and

$$f_2 = \frac{\alpha \beta^* (1 - \exp(i\Omega' (\vec{n} \vec{r})/c))}{\exp(-i\Omega' t) \left(\frac{\Omega' - \Delta}{\omega_p - \omega} + \frac{\Omega' + \Delta}{\omega_p - \omega - \Omega'} \right)},$$

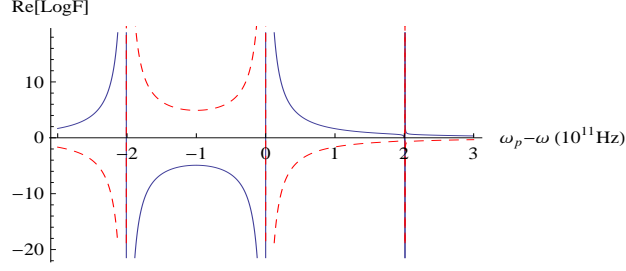


FIG. 1: (Color online) The real part of the exponent of prefactor F as a function of frequency difference of probe and pump fields at $t = \pi/\Omega'$ (solid line) and $t = 2\pi/\Omega'$ (dashed line). Other parameters are $z = \pi c/\Omega' \text{ cm}$, $\rho = 2 \cdot 10^{15} \text{ cm}^{-3}$, $\alpha = \sqrt{0.99}$, $\beta = 0.1$, $\Delta = -2 \cdot 10^{11} \text{ Hz}$, $\omega \approx 10^{15} \text{ Hz}$, $\omega_0 \approx 10^{15} \text{ Hz}$ and $\Omega = 2 \cdot 10^{10} \text{ Hz}$.

represents the contribution of red- and blue-sidebands, i.e. the superposition nature of the atomic state.

Expression of this prefactor $F(\vec{r}, t)$ is the main result of this paper. It has imaginary and real parts in the exponent, which mimics the respective parts of the bias current. The imaginary part of prefactor $F(\vec{r}, t)$ stipulates a phase modulation, while the real part introduces amplitude modulation and intensity variance of the probe laser beam during the propagation in the medium. On the other hand, the exponent is a periodic function of time and spatial coordinate, which results in a periodic-type modulation of the probe field intensity, accompanied by amplification or weakening in average. Fig.1 ascertains the situation in detail. The solid represents dependence of the real part of exponent on carrying frequency of probe field at the initial time moment (when the wavefront reaches the spatial coordinate z examined). Amplification or weakening of intensity depends on whether the frequency is in a region with positive or negative values of exponent (of first kind are left and right regions, of second kind – the intermediate one). The dashed line pictures the same dependence for half-period later. Situation is the opposite in this case: frequencies that were amplified now are decreasing and vice versa. Therefore, if for a given carrying frequency propagation through the medium starts from amplification, the probe field in the medium is an amplified periodic sequence of pulses, the minima values of which coincide with the entering value. If propagation starts from weakening, it gives the opposite regularity, namely, a sequence of pulses with maxima values repeating the entering value. A periodic flow of energy (photons) takes place between pump and probe fields. If the flow goes from pump to probe, then one gets the mentioned amplification (in average) and vice-versa in the opposite case.

Modulation periods $T_{\text{mod}} = 2\pi/\Omega'$ and $L_{\text{mod}} = 2\pi c/\Omega'$ are determined by the generalized Rabi frequency only, and can be widely tuned and easily controlled with high precision. The depth of modulation, besides the Rabi frequency and pump and probe frequencies, is determined by the product of gas density on a single photon scattering cross section. It is evident that deepening of modulation at a fixed value of repetition period T_{mod} or L_{mod} should result in narrowing of peaks in the modulation picture and formation of a periodic sequence of sharp (ultrashort) pulses. Simultaneously choosing the amplification regime, one gets just the desired comb of high power and ultrashort optical pulses.

The next point which should be brought to attention are zeros of denominators. In fact, they show processes that have contribution in intensity modulation (amplification and

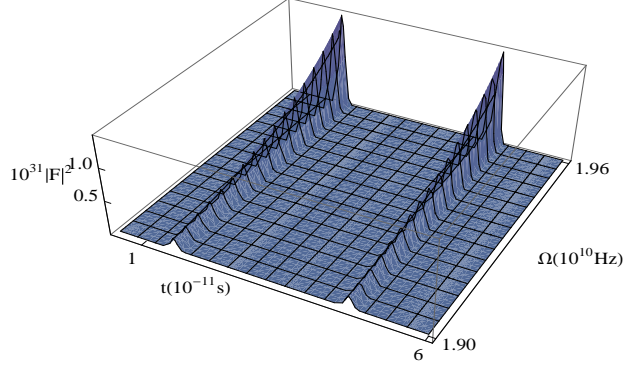


FIG. 2: (Color online) Modulation and amplification of the probe wave in two-level gaseous medium. Here $\omega_p - \omega = 2 \cdot 10^9 \text{ Hz}$, $\rho = 6 \cdot 10^{14} \text{ cm}^{-3}$ and the other parameters are as in Fig.1.

weakening). The condition $\omega = \omega_p$ corresponds to Rayleigh scattering while the other two at $\omega = \omega_p \pm \Omega'$ represent common atomic absorption/emission and Stokes (anti-Stokes) type stimulated hypercombination processes, where absorption (emission) of two pump photons is followed by emission (absorption) of a probe photon transferring atom from the lower (upper) energy level to the upper (lower) one.

And finally, exponent of the prefactor is proportional to the pump intensity Ω . This means that a simple (linear) superposition of two dressed quantum states does not make feasible the process of pulse splitting yet. Here superposition takes place by means of some nonlinear processes, relevant to the system.

To conceive the picture of probe wave modulation, let's consider the two-level model of alkali metal gases. The characteristic values of $|d|^2$ for dipole allowed transitions are around $2 \cdot 10^{-34} \text{ CGSE}$ and sample concentration can be varied in a wide range of $10^{12} - 10^{16} \text{ cm}^{-3}$. Typical line broadening is $10^7 - 10^8 \text{ Hz}$ and therefore the lowest allowed (in frame of this model) value for resonance detuning is $|\Delta| = 2\pi \times 10^8 \text{ Hz}$. A picture of probe wave modulation under some possible conditions is given in Fig.2. It shows the ability of the presented scheme of interaction to juxtapose the composition of high repetition ultrashort pulses (duration of an individual pulse is about 250 fsec) with their huge amplitude amplification, desirable for high-field light-matter interactions and implementation of high-precision comb-spectroscopy, for instance.

When modulation conditions are not satisfied, the probe wave is simply a running wave propagating in a dielectric medium. Expression (9) for $n_0(\omega)$ (gas refractive index) in comparison with earlier known result [12] contains out of dipole approximation term $(e^2/m)(\Omega^2/\Omega')$ in both rectangular brackets. It is interesting that contribution of this additional term is of resonant nature and has no saturation in dependence on pump wave intensity Ω . It repeats recently obtained regularity for the parametric down conversion process [13]. This out of dipole approximation contribution appears attractive in far IR or longer wavelength ranges of radiation. For example, in the range of the CO_2 laser operation (9.4 – 10.6 micrometers) its contribution is already about 10 %.

Note that our analytic treatment (in particular, presentation of medium dispersion properties in the frame of two-level approximation) is essentially grounded on the usage of slowly varying approximation and implies at least some tens of femtoseconds for laser light pulse duration

In conclusion, a consistent discussion of the propagation of off-resonance quasimonochromatic radiation, incident to a gaseous two-level medium prepared in a quantum superposition state of internal dressed states, shows that it is split into a sequence of ultrashort pulses with easy and precise tuning of possible control parameters. The split dynamics can be accompanied, in general, by large gain or loss in average. Being necessarily based on quantum nature of superposition in each individual atom, the observation of such modulation can be regarded as exhibition of a new quantum macroscopic optical phenomenon.

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